

## **Beamline 5.3.1**

# **Femtosecond Phenomena**

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# Femtosecond Synchrotron Pulses from the Advanced Light Source

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An important new area of scientific research in chemistry, physics, and biology is the investigation of ultrafast structural dynamics in condensed matter using femtosecond x-ray pulses. The dynamic properties of materials are governed by atomic motion which occurs on the fundamental time scale of a vibrational period,  $\sim 100$  fs. This is the time scale of interest for ultrafast chemical reactions, non-equilibrium phase transitions, vibrational energy transfer, surface desorption and reconstruction, and coherent phonon dynamics. To date, our understanding of these processes has been limited by lack of appropriate tools for probing the atomic structure on an ultrafast time scale. X-ray techniques such as diffraction and EXAFS yield detailed information about “static” atomic structure. However, the time resolution of high-brightness synchrotron x-ray sources such as the Advanced Light Source ( $\sim 30$  ps) is nearly three orders of magnitude too slow to directly observe fundamental atomic motion. Conversely, femtosecond lasers measure transient changes in optical properties of materials on a 10 fs time scale, but optical properties are only indirect indicators of atomic structure.

We have recently demonstrated a novel scheme for generating ultrashort pulses of synchrotron radiation[1]. Our approach is to create femtosecond time-structure on a long electron bunch by using a femtosecond laser pulse to modulate the energy of an ultrashort slice of the bunch. The modulation is achieved via interaction between the electrons and the light field in a wiggler (Figure 1a). Half of the electrons overlapping with the ultrashort pulse are accelerated and half are decelerated depending on the optical phase. The laser-induced energy modulation is several times larger than the rms beam energy spread at the ALS ( $\sim 1.8$  MeV), and only an ultrashort slice of the electron bunch experiences this modulation. The accelerated and decelerated electrons are then spatially separated from the rest of the electron bunch (in a dispersive bend of the storage ring) by a transverse distance that is several times larger than the rms transverse size of the electron beam (Figure 1b). Finally, by imaging the displaced beam slice to the experimental area, and by placing an aperture radially offset from the focus of the beam core, we will be able to separate out the radiation from the offset electrons (Figure 1c).

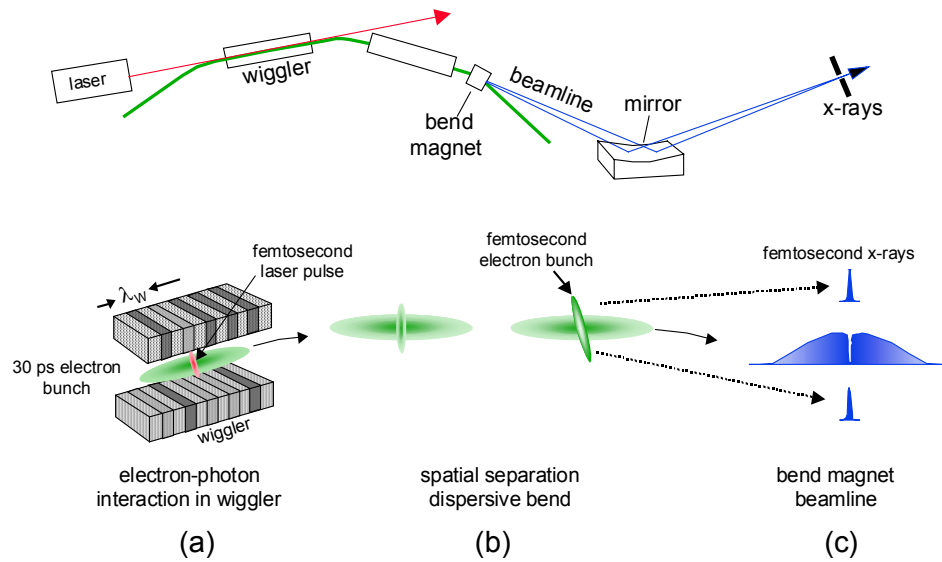


Figure 1. Schematic illustration of the technique for generating femtosecond synchrotron pulses. (a) laser/electron beam interaction in resonantly-tuned wiggler, (b) separation of accelerated femtosecond electron slice in a dispersive (bend) section, (c) generation of femtosecond x-rays at a band-magnet beamline.

This technique has recently been implemented on ALS beamline 5.3.1. A cryogenically cooled Ti:sapphire femtosecond laser system is located near the beamline endstation, and the laser pulses ( $\sim 1$  mJ, 100 fs pulse duration, 2 kHz repetition rate) are projected across the storage ring roof blocks to sector 5, where they enter the main vacuum chamber through a back-tangent optical port. Amplified femtosecond pulses co-propagate with the electron beam through wiggler W16 in sector 5. A photon stop/mirror following the wiggler reflects the laser light and the visible wiggler emission out of the vacuum chamber for diagnostic purposes in order to verify the spatial, spectral, and temporal overlap of the laser beam with the fundamental (undulator-like) emission from the wiggler.

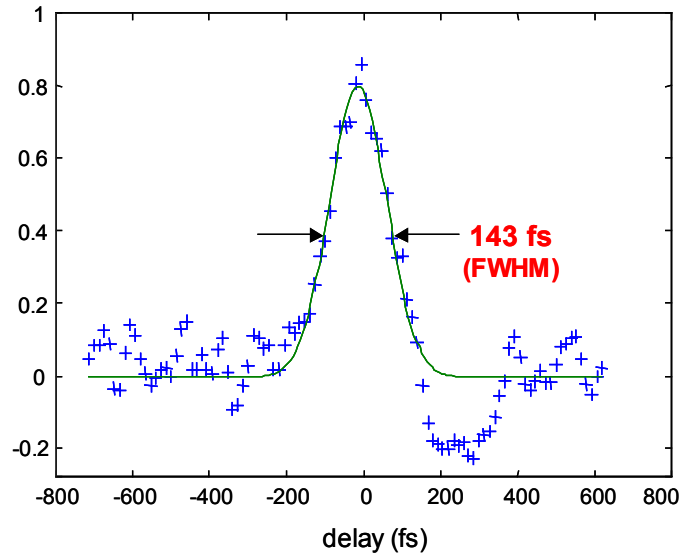


Figure 2. Cross correlation of an ultrashort synchrotron pulse with femtosecond laser pulse. The deconvolved synchrotron pulse duration is 143 fs (FWHM).

Femtosecond duration synchrotron pulses are directly measured by cross-correlating the visible light from bend-magnet beamline 5.3.1 at the ALS with a delayed laser pulse in the nonlinear crystal, BBO. Figure 2 shows a laser synchrotron cross-correlation measurement corresponding to synchrotron pulses of <150 fs duration, the shortest pulses ever generated from a synchrotron. Additionally, these measurements were made during normal multi-bunch operation of the storage ring, which illustrates the compatibility of this approach with the requirements of other synchrotron users, notably the protein crystallography beamline which simultaneously uses the wiggler W16. An important point is that the femtosecond time structure will be invariant over the entire spectral range of bend-magnet emission from the near infrared to the x-ray regime, making this a very powerful tool for femtosecond spectroscopy.

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# Fs x-ray measurement of short-range structural dynamics in VO<sub>2</sub> during a photo-induced phase transition.

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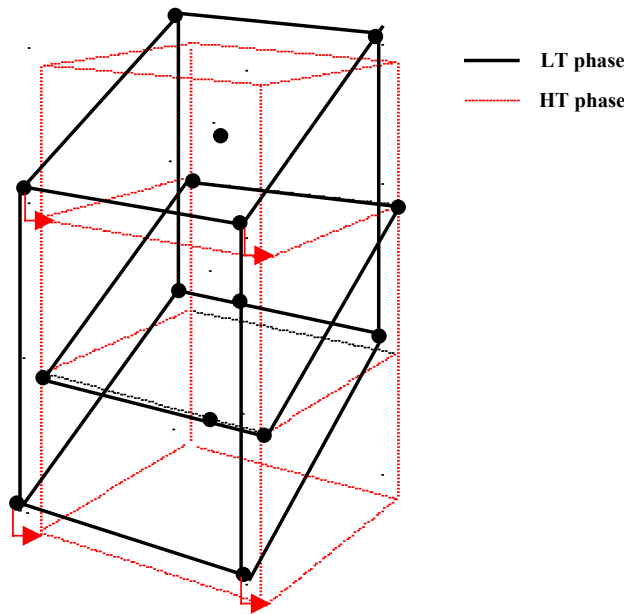
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## INTRODUCTION

The main goal of this experiment is the measurement of femtosecond structural and electronic dynamics during a phase transition in the correlated, non-magnetic oxide VO<sub>2</sub>. This material undergoes a metal-insulator transition at 340 K, accompanied by a structural transformation between a low-T monoclinic and a high-T rutile phase. The role of electron-electron and electron-phonon correlations during this process has long been debated<sup>1</sup>. Current understanding favors the picture of a Mott-Hubbard insulator (low-T phase), which is turned into a simple metal upon relaxation of a structural distortion above the transition temperature.



**Fig. 1.** Structural transition between the low-T monoclinic and high-T rutile phases of Vanadium Dioxide. A transverse optical phonon drives the transition within less than 1 picosecond.

Recent femtosecond x-ray diffraction measurements<sup>2</sup> have indicated that the structural transition has a very fast component, whereby long-range order between Vanadium atoms is changed within less than one picosecond. Static Raman spectra suggest that this structural transition may be impulsively induced, by photo-excitation of transverse optical phonons involving Vanadium-Vanadium distortions<sup>3</sup>. Further, while an additional strong V-O Raman-active mode is found (not shown in the figure) it is also known that a prominent signature of the metal insulator transition can be found near the Oxygen absorption edge<sup>4</sup>, due to the strong hybridization of p and d orbitals from the Vanadium and Oxygen atoms, respectively.

## CURRENT STATUS

The correlation between excitation of these vibrational modes and ultrafast changes in the electrical properties is the main object of our study. The experiments require combined measurement of the ultrafast electrical transition (by means of femtosecond NEXAFS spectroscopy) and of short-range structural dynamics, with sensitivity to both light (Oxygen) and heavier (Vanadium) elements (by means of femtosecond EXAFS).

The experiment will be performed using femtosecond pulses of synchrotron radiation available at the Advanced Light Source, in the spectral range between 500-800 eV, i.e. across the Vanadium L-edges and the Oxygen K-edge. The feasibility of these measurements has been demonstrated in grazing incidence geometry, where signatures of both phase transitions (electrical and structural) have been identified. For this purpose, we have performed static experiments (by varying the temperature of the sample), and time-resolved pump-probe experiments in the hundred picosecond timescale.

Because of the inconvenience of grazing incidence geometry, current efforts are dedicated to the fabrication of thin (0.05  $\mu\text{m}$ ), free-standing films of Vanadium Dioxide, using a combination of deposition and etching techniques. In the future, direct measurements of the EXAFS and NEXAFS component of the  $\text{VO}_2$  transition will be conducted in a transmission geometry.

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# Highly sensitive XAS with Single X-Ray Pulses at 1 kHz

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## INTRODUCTION

We are currently developing a scheme to exploit pulsed x-radiation for structural dynamics studies at beamline 5.3.1 via time-resolved X-ray absorption spectroscopy (XAS). While XANES is complicated to interpret from a structural point of view, it contains rather simple phenomena like the chemical shift, which describes the oxidation state of the atom. On the other hand, EXAFS delivers a detailed picture of the local environment [1], and interatomic separations can nowadays be determined with an accuracy down to 100 fm [2]. In addition, EXAFS requires no periodic structures (as required in x-ray diffraction), and can be readily applied to disordered systems, e.g., liquids.

Laser-pump SR-probe experiments depend on the available number of x-ray photons per single pulse at the repetition rate of the exciting laser, which is typically 1 kHz for commercial amplified fs-lasers. Therefore, such experiments can use only about  $10^{-5}$  -  $10^{-6}$  of the available photon flux at a synchrotron with its much higher pulse repetition rate of 100 - 500 MHz. We have performed calculations of the expected pump-probe signal for a given condensed phase chemical system [3,4], which also underline the general utility of time-resolved XAS in condensed phase dynamics research.

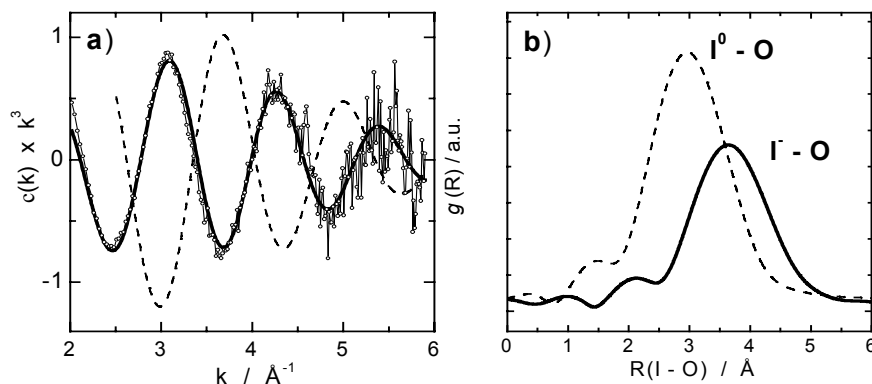
## EXPERIMENTAL APPROACH

X-radiation from the ALS camshaft pulse (detected with an APD behind the sample) is singled out with a gated integrator (opening window ca. 20 ns). In order to record high-quality spectra in a pump-probe configuration we have employed the following scheme: The gated integrator is triggered at twice the laser repetition rate, so that every second x-ray pulse corresponds to the static (= unexcited) sample, while the other x-ray pulses monitor the transmission following photoexcitation at an adjustable time delay. Therefore, the gated integrator delivers an output of alternatively laser-pumped and unpumped intensities, which are then read into the computer and appropriately sorted. With this scheme we can record high-quality EXAFS spectra of the photoinduced changes. Indeed, a careful analysis of the measured noise shows that we have nearly achieved the shot noise limit in this configuration. The measured noise is considerably reduced over other methods, which include additional systematic noise sources. The step-scanning monochromator induces vibrations, which result in a rather constant noise source on the transmitted intensity. This is considerably reduced, when no optics move, and the decreasing storage ring current is observed by a corresponding broadening of the pulse height distribution. In addition, using our latest development by measuring the difference spectra *during* a monochromator scan reduces the noise even more. After correcting the statistically derived effective flux for the limited x-ray absorption in the 30  $\mu\text{m}$  thick APD, our measurements are very close to the specified flux of this beamline [5].

## RESULTS AND DISCUSSION

With our new difference-signal detection scheme we make best use of the x-ray source, since the recorded noise on these spectra correspond nearly entirely to the expected shot noise limit. But we can also record high-quality EXAFS without employing the difference measurement technique. Figure 1 illustrates this for the case of aqueous iodide. The energy spectrum has already been transformed into  $k$ -space (Fig. 1a), but the original data has not been treated (e.g., smoothed or Fourier-filtered). This spectrum represents a single scan accumulating 2500 single x-ray pulses per data point. The fit (and its Fourier transform, Fig. 1b) delivers a nice agreement with the literature, and the accuracy for determining the nearest neighbor shell of oxygen atoms is better than 10 pm [4].

Fig. 1 also illustrates the potential for measuring the nearest-neighbor distance in photogenerated iodine radicals. The I-O distance should change considerably upon photodetachment. Values for iodine are currently unknown, but in a theoretical study of aqueous Br radicals the predicted Br-O distance changed from 335 pm for the ion to ca. 280 pm for the neutral radical [6]. Assuming a similar reduction in the iodine-oxygen distances would result in a pronounced change in the EXAFS signal, as shown in the dashed curves in Fig. 1.

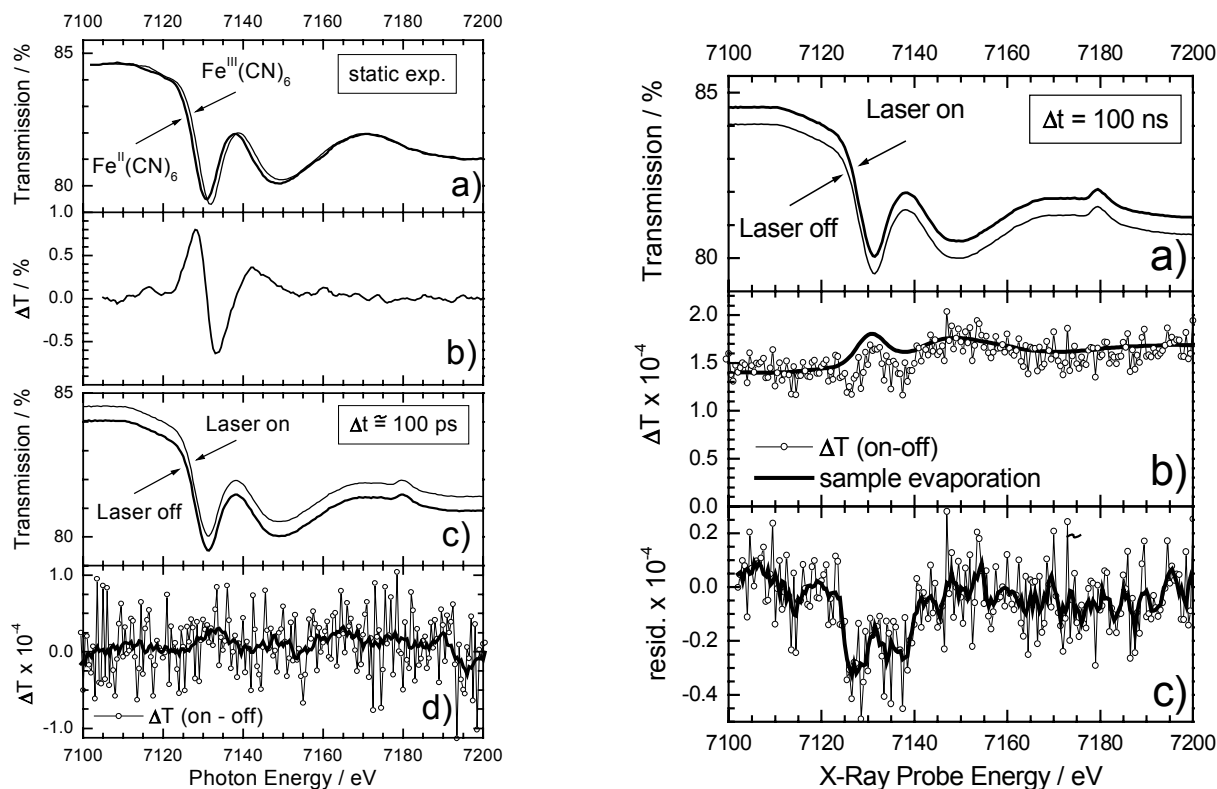


**Figure 1.** Static EXAFS above the  $L_3$  edge of iodine of aqueous iodide (NaI concentration: 0.7 mol/l, thickness: 0.1 mm). Original data (open circles), and their fit (solid line) is shown in a), and the Fourier transform showing the nearest neighbor I-O distance in b) (solid line). For comparison, a simulation of the expected EXAFS of atomic iodine is shown (dashed curve in a) together with its Fourier transform (dashed curve in b)).

Exploiting our difference measurement technique we have attempted to measure the photo-induced chemical shift due to UV photoionization of the central Fe atom in aqueous  $[\text{Fe}^{\text{II}}(\text{CN})_6]^{4-}$  with a synchronized fs laser. The chemical shift of ca. 1 eV around the Fe K edge was observed in static samples containing either  $[\text{Fe}^{\text{II}}(\text{CN})_6]^{4-}$  or  $[\text{Fe}^{\text{III}}(\text{CN})_6]^{3-}$  (concentration 0.2 mol/l, sample thickness 0.1 mm), as illustrated in Fig. 2 a) and b). Then we excited a liquid jet with the reactant species  $[\text{Fe}^{\text{II}}(\text{CN})_6]^{4-}$  only with 100  $\mu\text{J}$  of 266 nm light, and recorded the transient changes. Due to the low excitation yield of below 0.4 % we could not observe the chemical shift on the 100 ps time scale (Fig. 2 c) and d). However, with improved statistics exploiting the first 100 ns of the multibunch train following photoexcitation we did observe a photoinduced change in the XAS. In Fig. 3 a) the average of 20 spectra are shown for each the unpumped and the pumped sample (offset for clarity). Fig. 3 b) shows the difference signal of Fig. 3 a) together with a simulation assuming laser-induced evaporation of ca. 100 nm of the 0.1 mm thick sample. While this nicely reproduces the majority of the measured difference spectrum, we still observe some discrepancy around the  $1s \rightarrow 4p$  pre edge feature, as shown in the residual spectrum in Fig. 3 c). The double minimum shape of decreased transmission (clearly visible in the smoothed spectrum) indicates a



broadening of this feature, which has been observed in temperature-dependent XANES studies. The elevated temperature of the laser-heated sample could account for this observation. While our current sensitivity does not allow to unambiguously observe the chemical shift for less than 0.4 % excited species, current improvements on the laser excitation process will permit us to boost the product yield into the 1-5 % range.



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# Light Induced Intersystem Crossing in $\text{Fe}[(\text{py})_3\text{tren}](\text{PF}_6)_2$ Crystals

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## INTRODUCTION

$\text{Fe}[(\text{py})_3\text{tren}]^{2+}$  is a member of a class of octahedrally coordinated iron-based transition metal complexes which undergoes an  $\Delta S=2$  spin-crossover transition upon light excitation. This transition proceeds first by electronic excitation from a low-spin singlet ground state to a metal-to-ligand charge transfer state, followed by subsequent relaxation to a high-spin quintuplet state, a process completed within roughly 500 femtoseconds at room temperature. The lifetime of the excited, high-spin state is roughly 700 picoseconds at room temperature, after which the system relaxes back to the singlet ground state. Concomitant with this highly spin-forbidden electronic transition is a structural dilation of the ligand cage surrounding the iron. Current experimental work has only been able to establish a symmetric dilation of the ligand cage in the high-spin state, with the bonds of the first shell of octahedrally-coordinated nitrogen distended by 10% relative to the low-spin state. It has been conjectured, however, that in the intermediary stages of the spin-crossover process, the ligand cage, in fact, distorts asymmetrically, and that it is this distortion which gives energetic preference to the high-spin state. With the ultrashort x-ray pulse capabilities of beamline 5.3.1, we hope to observe dynamically these structural changes in the iron's ligand cage to provide evidence of the role, if any, of the structural changes in the spin-crossover behavior of  $\text{Fe}[(\text{py})_3\text{tren}]^{2+}$ .

## CURRENT STATUS

We intend to analyze EXAFS and near-edge structures of the iron and nitrogen to determine the atomic motion of the ligand cage as the system evolves after excitation. Due to the signal-limited nature of this experiment, a great deal of effort has gone into the design and characterization of the measurement apparatus. Offline, work has focused on crystal synthesis and sample preparation to provide for optimal signal-to-noise, and optical characterization of the dynamics to determine optimal excitation conditions.

Static spectra of the iron K-edge have been recorded for the candidate molecule and a chemically synthesized high-spin analogue to anticipate the potential spectral changes that may be observed between the two states. These time-independent spectra are shown in Fig. 1 and clearly demonstrate a near-edge shift between the high and low spin states. We intend to next observe similar spectral shifts in a time-resolved fashion following excitation by femtosecond laser pulses.

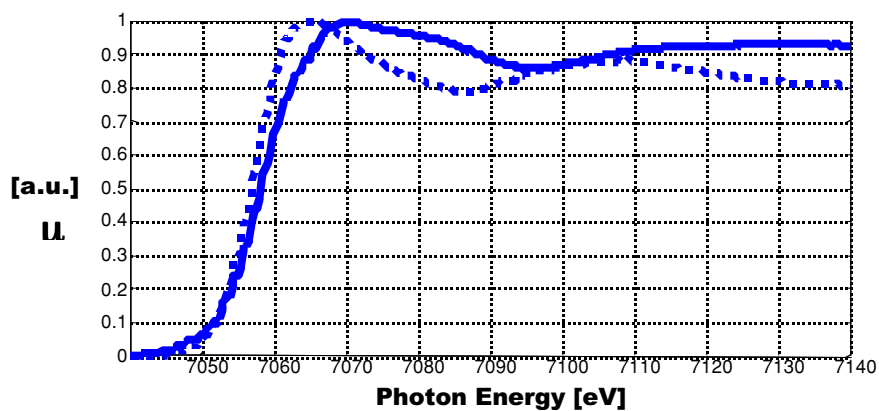


Figure. 1 Static EXAFS spectra of high (dashed line) and low (solid line) spin states of  $\text{Fe}[(\text{py})_3\text{tren}]^{2+}$ .

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# Ultrafast Measurements of Liquid Carbon and Liquid Silicon

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## INTRODUCTION

Conventional techniques of x-ray absorption spectroscopy, while extremely useful for studying the electronic and atomic structures of many materials, are generally not suitable for high temperature, volatile materials. The introduction of time resolution overcomes much of this difficulty: by melting thin foils with an ultrafast laser pulse and measuring the transmission of x-rays before break-up of the liquid, we can obtain useful absorption spectra of high-temperature liquids.

## EXPERIMENT AND RESULTS

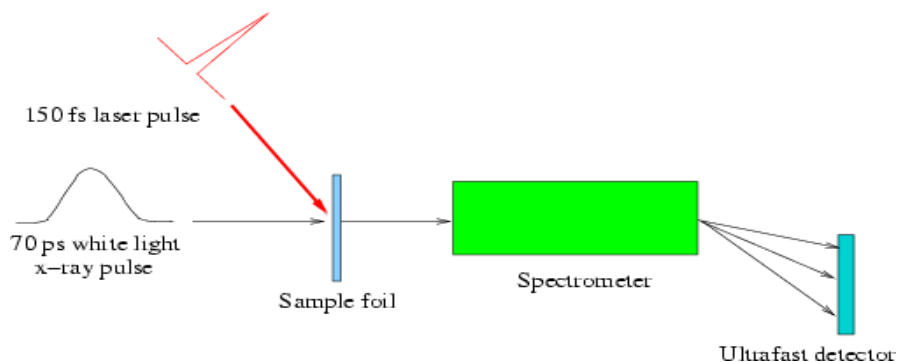


Figure 1. Sketch of experimental setup for time-resolved absorption.

Figure 1 shows a sketch of the experimental setup at beamline 5.3.1 of the Advanced Light Source. After heating the foil with an 800 nm, 150 femtosecond laser pulse, a 70 ps pulse of broad spectrum x-rays from a synchrotron bend magnet pass through the laser-excited region of the foil. A spectrometer then disperses the x-rays onto a detector. The detector is either a set of microchannel plates (for pump-probe measurements with 70 ps time resolution) or an ultrafast x-ray streak camera (for a resolution of up to 1 ps).

Figure 2 compares the  $L$ -edge spectrum of an unheated silicon foil with the spectrum of the foil 100 ps after laser excitation. The transition from solid to liquid results in a number of changes to the spectrum: a 50% drop and 2 eV broadening in the  $L_{II,III}$  edge at 100 eV, a  $-1.6 \pm 0.2$  eV shift in the  $L_I$  edge at 150 eV, and a dramatic decrease in the magnitude of EXAFS oscillations. Model calculations based on molecular dynamics simulations of liquid silicon compare favorably with these results.

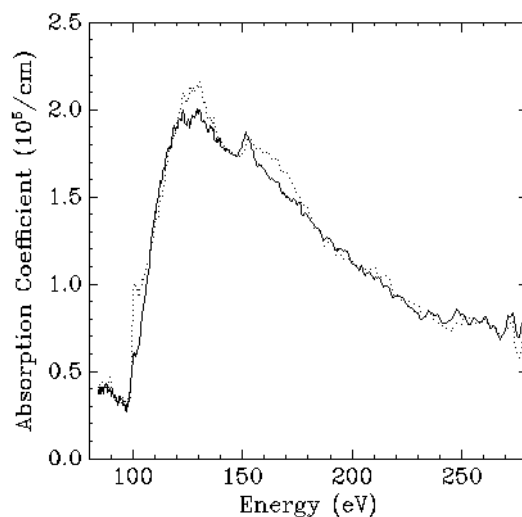


Figure 2. Absorption Spectra of silicon L-edges. Dotted line: unheated silicon; Solid line: heated silicon, 100 ps after laser.

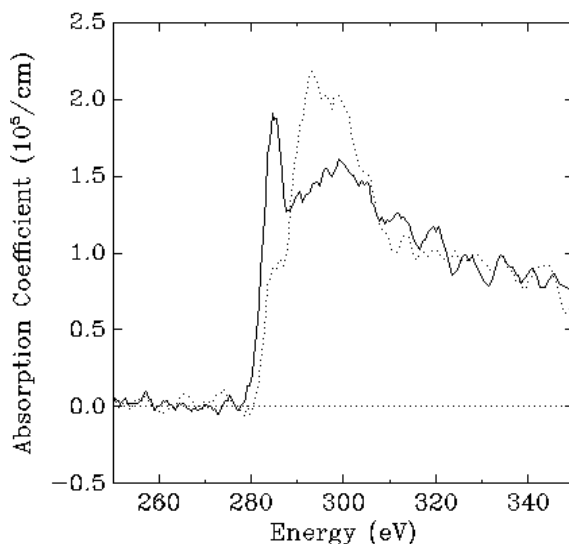


Figure 3. Carbon K-edge absorption spectra. Dotted line: unheated carbon; solid line: heated carbon, 100 ps after laser.

Measurements on foils of soft amorphous carbon, summarized in figure 3, show large differences between the unheated and heated spectra of the carbon *K*-edge. After 100 ps, the laser-induced melting causes an increase in the size of the  $\pi^*$  resonance at 285 eV accompanied by a decrease and 4 eV shift in the  $\sigma^*$  resonance at 296 eV. These changes indicate a transition in bonding geometry from the predominant  $sp^2$  bonding characteristic of soft amorphous carbon to the collection of distorted  $sp$  bonded chains recently predicted by molecular dynamics simulations for the low-density liquid phase of carbon.<sup>1</sup>

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